AEROSOL MANUFACTURING TECHNOLOGY FOR PRODUCTION OF LOW- COBALT LI-ION BATTERY CATHODES

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Project ID# BAT411

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OVERVIEW

Timeline

- Project start date: 10/01/2018
- Project end date: 06/30/2022
- Percent complete: 65%

Budget

- Total project funding: $3,749,057
  - DOE share: $2,989,057
  - Contractor share: $ 760,000
- Funding for BP1 (2018-19): $1,133,905
- Funding for BP2 (2020-21): $1,369,863
- Funding for BP3 (2021-22): $1,245,289

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Barriers and Technical Targets

- Performance: cell chemistries that provide higher energy have life and performance issues
- Life: next gen technologies still suffer major cycle and calendar life issues
- Cost: main drivers are the high cost of raw materials and materials processing, the cost of cell and module packaging, and manufacturing costs

Partners

- ANL: Joseph Libera – spray pyrolysis process development, Eungje Lee- cathode development/characterization
- SAFT: Carine Margez, Serena Peterson – cell components, fabrication and testing

RELEVANCE - PROJECT OBJECTIVES

Impact:

- Address both cost and performance challenges for the next generation Li-ion batteries (LIB). If successful, the project will have significant impact on cost reduction of Li-ion battery towards the $100/kWh target
- Flexibility to produce all key Low-Cobalt cathode compositions

Objective:

Research, develop, and demonstrate aerosol manufacturing processes to produce of low/free Cobalt active cathode materials for next-generation Li-ion batteries (LIB) capable of achieving the following performance targets:

Beginning of Life Characteristics at 30°C	Cell Level	Cathode Level
Useable Specific Energy @ C/3		≥600 Wh/kg
Calendar Life (< energy fade)	15 Years	
Cycle Life (C/3 deep discharge <20% energy fade)	1,000	
Cobalt Loading	≤ 50mg/Wh	
Cost	≤ $100/kWh	

APPROACH/STRATEGY

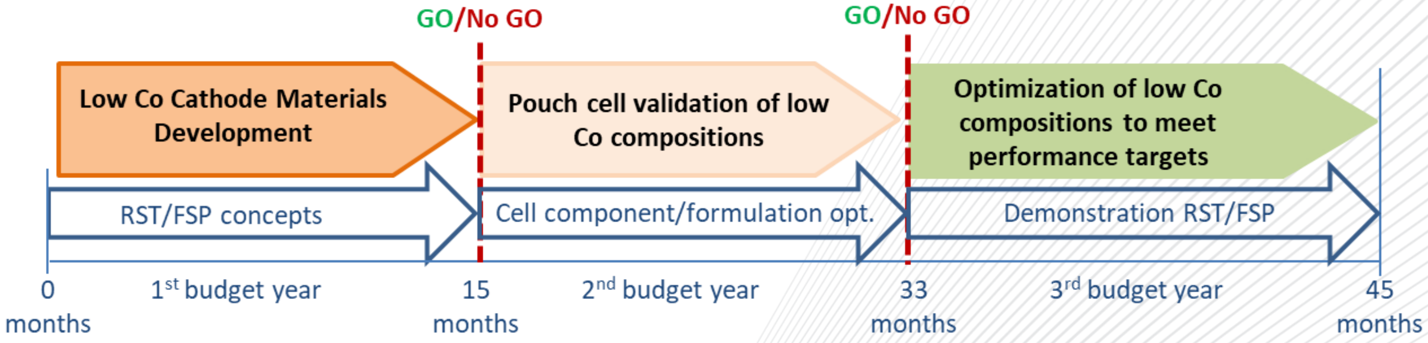
How to enable low/free Cobalt cathodes and improve cost and performance?

1. Materials supply chain optimization
   - Access to metals, strategic collaborations
2. Materials Improvements
   - Compositional modifications (less Co, cheaper raw materials)
   - Surface modifications (coatings, core/shell)

3.Improved synthesis process
   - Often as important as composition
   - Industry needs new ideas and approaches

Approach

- Develop Low-Cobalt cathode materials via Reactive Spray Technology (RST) and Flame Spray Pyrolysis (FSP) targeting < 50 mg_{Co}/Wh
- Implement structural and morphological modifications through aerosol particle production process
- Synthesize high Ni content NCMs, disordered rock-salt and other free-Co materials
- Optimize conductive additive formulations for Low/free Co cathode active material



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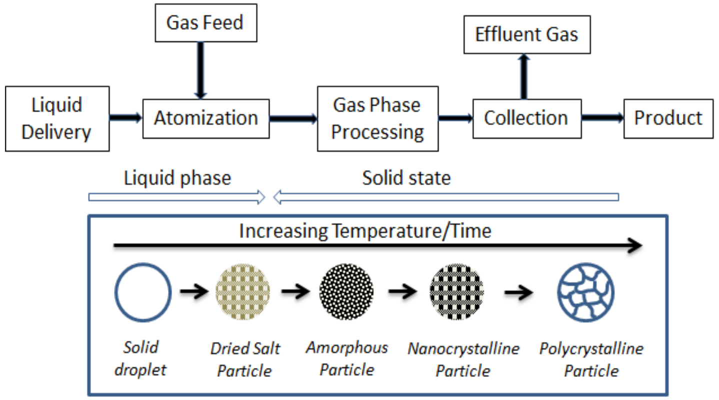
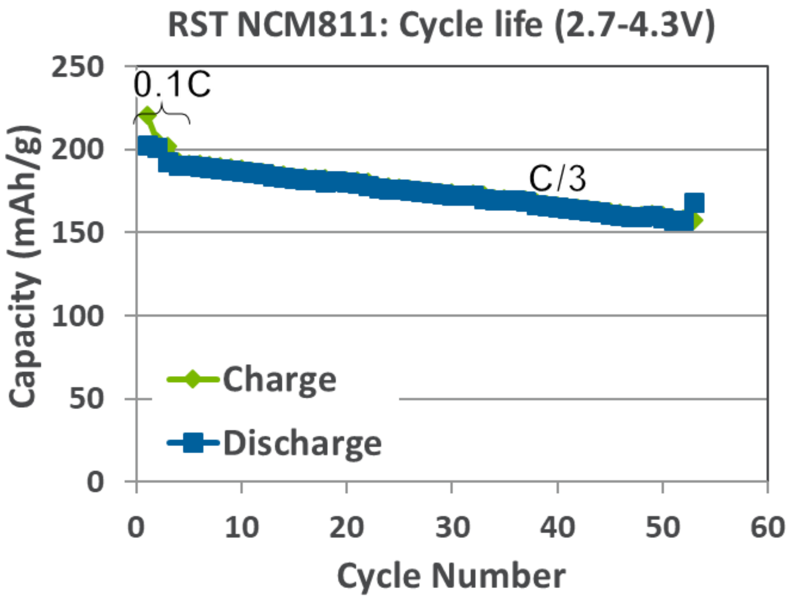
SPRAY PYROLYSIS PROCESS OFFERS COST AND SCALE BENEFITS TO PRODUCE LOW-CO CAMs

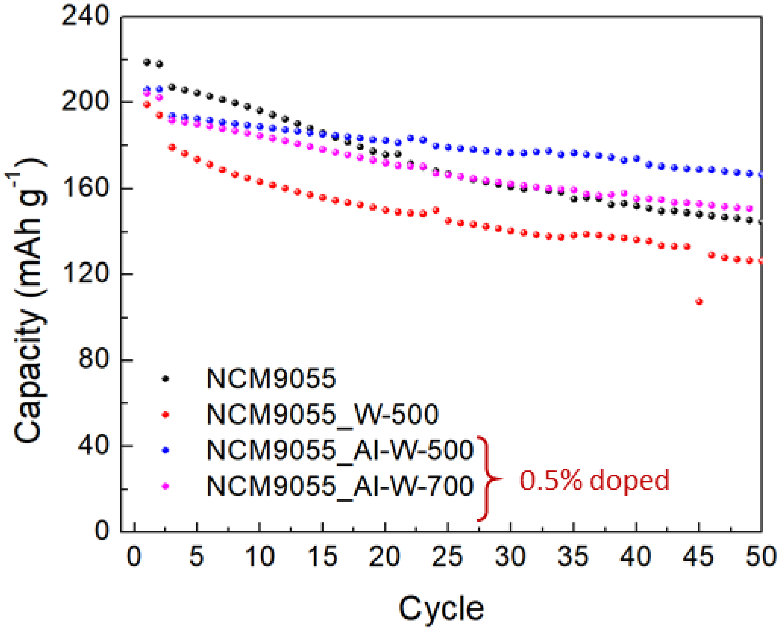
Reactive Spray Technology (RST)

- Continuous process simplifies/ eliminates heat treatment
- Liquid precursors: compositional flexibility and homogeneity, control particle density
- Fine spray/dry feed for small and controllable particle size
- Broader range of particle synthesis: micron-size containing nano-domains
- ***Hi-Ni content NCMs (811, 9055) and other low-Co compositions like NCA and NC946 have been produced***

RST system at Argonne National Lab (MERF)

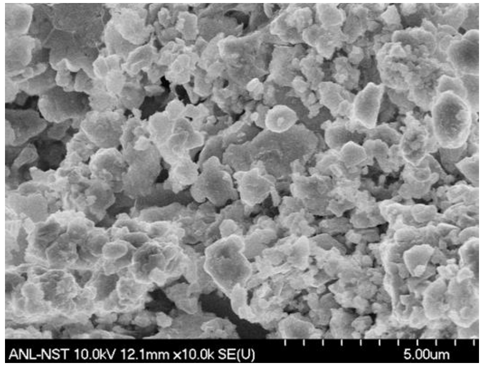


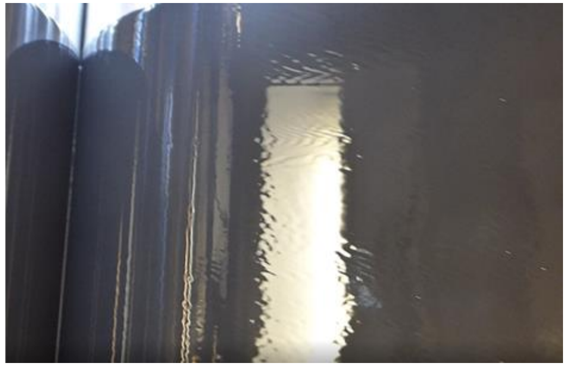




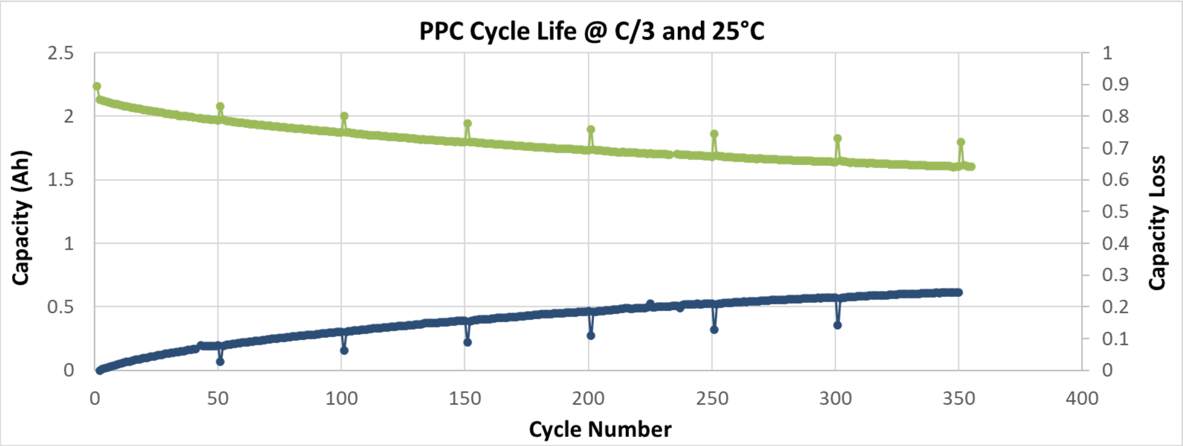
RST NCM811 WAS USED TO BUILD PROJECT PROGRESS CELLS (PPC)

- > 4kg of NCM811 were synthesized and postprocessed (ANL)
- Forty-five 2.1 Ah cells were assembled, tested and shipped to INL
- Slurry quality, electrode fabrication and cell performance checks followed industrial and USABC protocols
- PPC cycle life testing (SAFT) shows encouraging results (*no surface coating was applied to NCM811 particles*)



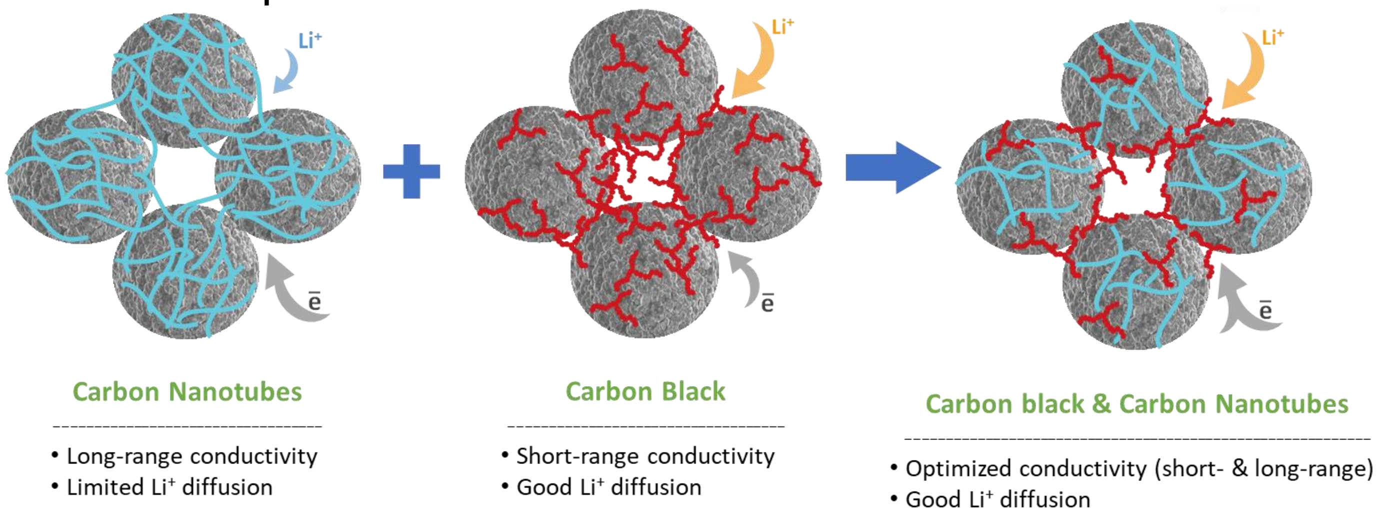


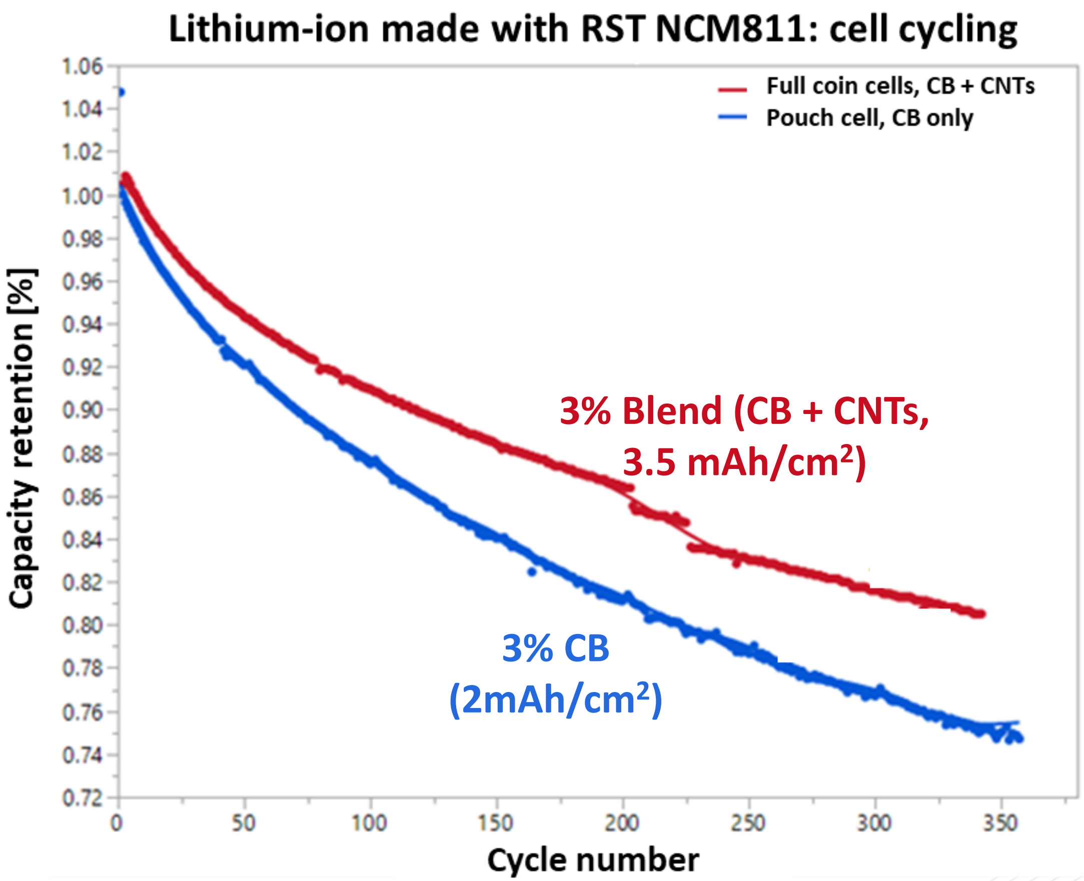
NCM811 Sample	Sample (g)	pH	Li_2CO_3 (wt%)	LiOH (wt%)	Engineering Decision
50-g batch	4.0223	11.4	0.24	0.17	---
PPC campaign	4.0072	11.4	0.17	0.14	Ok to process
Reference	2.0031	11.2	0.050	0.16	Commercial 811



CNTS/CB BLENDS IMPROVE HI-NI NCM ELECTRODE PERFORMANCE

- ***Optimize carbon conductive additive (CCA) formulations*** for Low/free Co active material cathodes to further increase performance
- ***Blends of carbon black (CB) and carbon nanotubes (CNTs) to improve charge transfer***
  - CNTs provide long range conductivity, but limited Li+ diffusion
  - Porous CB offers better Li-ion diffusion than CNTs
  - CB helps to mitigate poor ion transfer occurring at low temperature



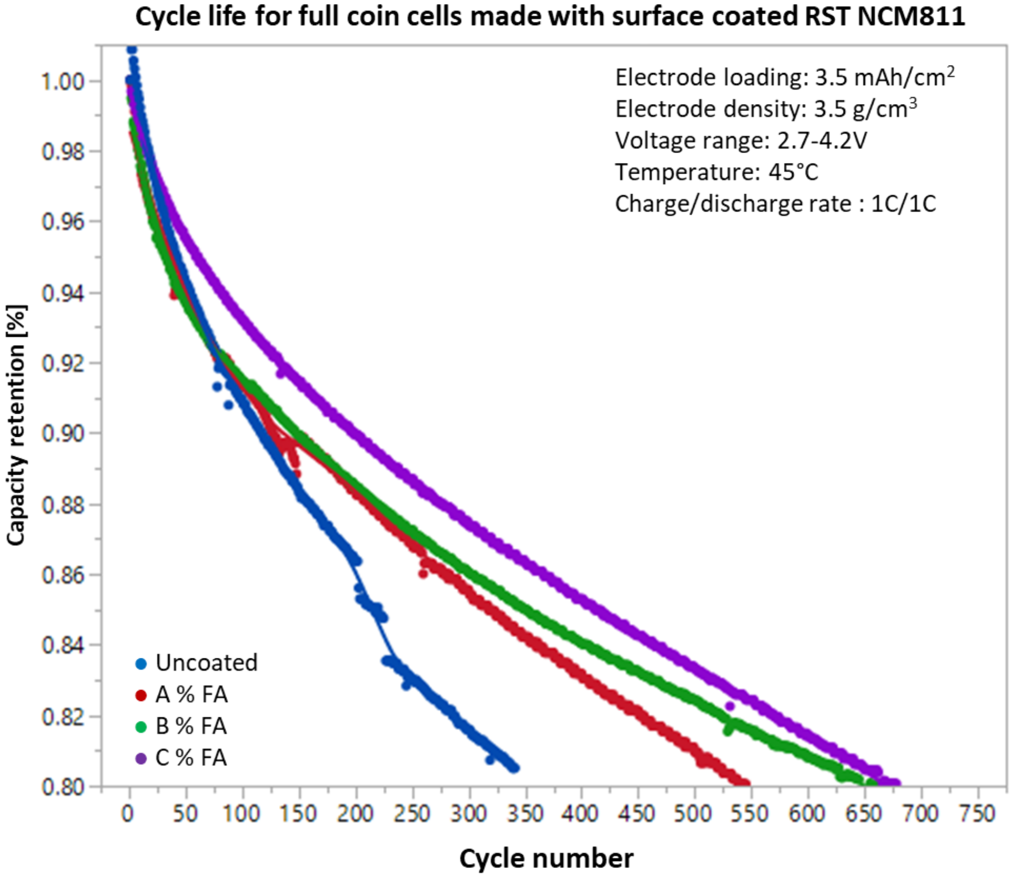


There is a clear improvement of cycling performance by using Cabot's CCA Blend

FUMED ALUMINA SURFACE COATING ENHANCES NCM's CYCLE LIFE

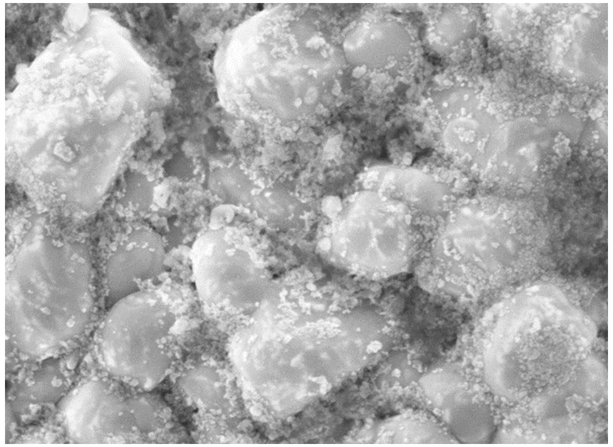
Dry coating

- Process: powder mixture + heat treatment
- Custom designed nanoparticles: simple metal oxides (i.e., Al_2O_3) and mix-metal oxides (LiM_xO_2)

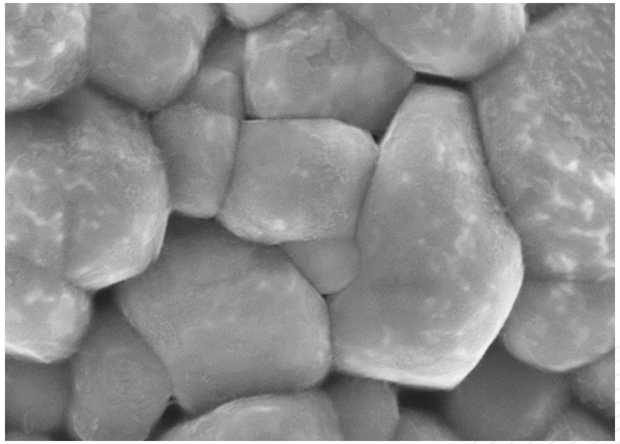


Fumed alumina (FA) surface modified NCM811

- Electrodes: CCA Blend, 3.5m Ah/cm^2
- Different amounts of FA: C > B > A > 0.2 wt%
- FA creates a more stable particle surface layer boosting capacity retention, while suppressing impedance growth



FA dispersed on NCM811 before heat treatment



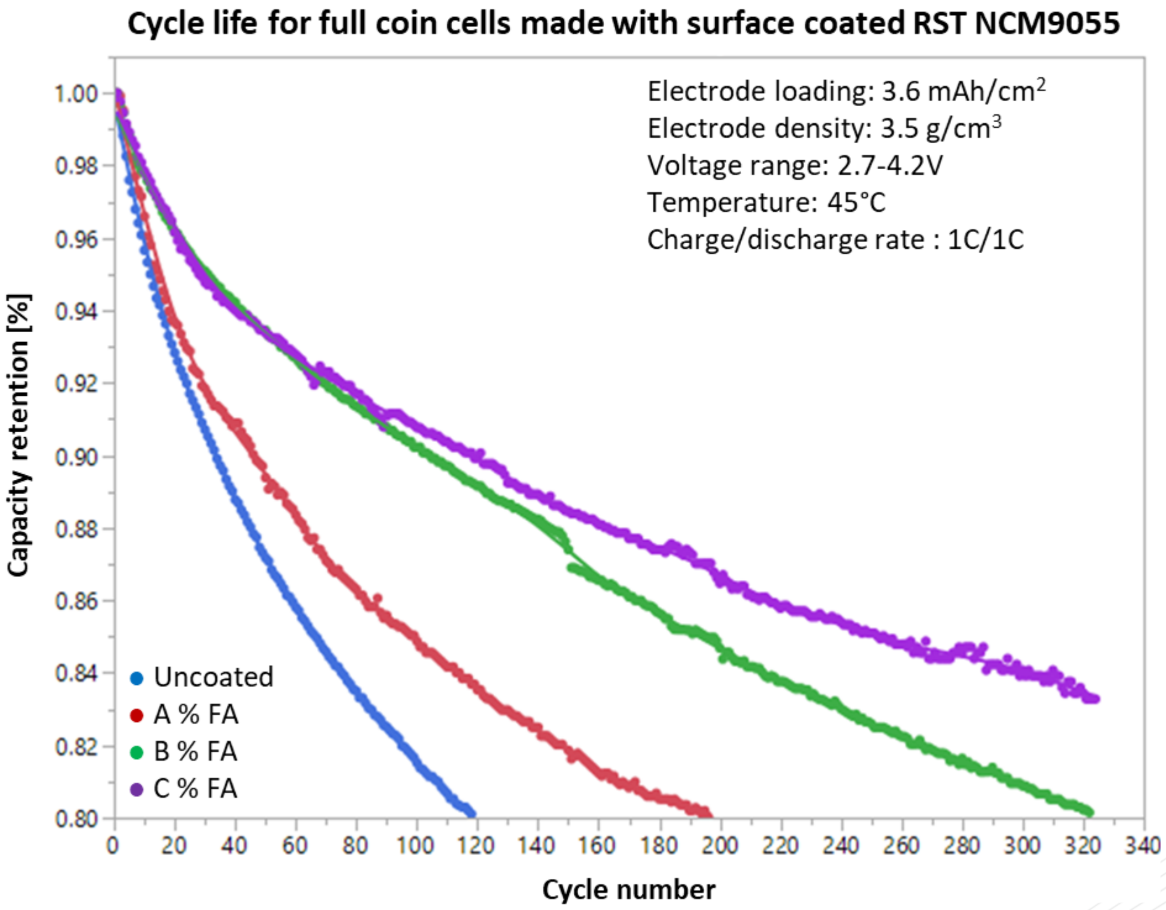
FA-surface coated NCM811After thermal treatment

FA SURFACE COATING BOOSTS NCM9055 ENERGY RETENTION

- Different wt% FA were applied to the surface of NCM9055 particles
- Electrodes: CCA Blend, 3.6 mAh/cm^2
- FA coating reduces impedance growth during high temperature storage and cycle life
- Leading to enhanced capacity and energy retention during cycling

Active material (SoC)	Cathode Energy Density (Wh/kg) 0.1D initial 100%	0.1D initial 80%	400 cycles, @ 0.1D 100%	400 cycles, @ 0.1D 80%
Un-coated NCM811, CB [1]	660	623	505	474
FA-coated NCM811, CCA Blend [2]	650	618	532	515
FA-coated NCM9055, CCA Blend [2]	675	649	556	532

*Cells were cycled at 1C/1C and 45°C, [1] Pouch cells, [2] Full coin cells



- **Surface coating and optimized CCA have improved NCM9055 performance**
- **Next: Interim pouch cells built using FA coated NCM9055, CCA blend and improved electrolyte**

COLLABORATIONS

- Argonne National Laboratory (ANL): Cathode compositions, cathode synthesis (spray pyrolysis), synthesis know-how, battery and analytical capabilities
  - ANL team has been working on Li-ion cathodes and other battery materials for more than 15 years. They have extensive understanding of cathode materials, electrodes, and unique facilities for the fabrication, characterization and testing of battery materials.
  - ANL team has developed high performance LIB cathode compositions such as layered-layered high energy materials with extremely high capacity.
- SAFT: Battery design, fabrication and testing; will assist selecting battery component materials
  - World leader in providing Li-ion systems for commercial, defense and space markets.
  - Experience manufacturing pouch cells ranging from 2Ah to 50Ah.
- STA Technologies/SICPA: collaboration to scale up spray pyrolysis production of powders, and assist to create a process cost model

PROPOSED FUTURE WORK

Budget Period 3 (2021-22)

- Optimize Low-Co cathode compositions, final refinement of FSP/RST cathode and powder production for project completion cells (PCCs)
- Select final surface modification process/composition for PCC selected cathode active material
- Optimize electrode formulation and architecture: finalize the design of cathode and selection of other cell components
- PCC build : fully optimized final battery design to build cell deliverables
- Deliver/test 30 PCC to INL (20) and TARDEC (10) in accordance with DOE cell testing protocols
- Any proposed future work is subject to change based on funding levels

End of project goal:

- Cell performance: > 600 Wh/kg (cathode) after 1000 cycles
- Cathode active material with ≤ $5mg_{Co}$/Wh

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SUMMARY

Relevance

- Address cell cost (≤ \$100/kWh) and cathode performance (≥ 600Wh/kg) challenges for the next generation LIBs
- Flexibility to produce all key low-Cobalt cathode compositions

Approach

- Develop low-Co cathode materials via RST and FSP targeting < 50 mg_{Co}/Wh
- Implement structural/morphological modifications through aerosol particle production process
- Synthesize high Ni content NCM and disordered rock-salt materials
- Optimize conductive additive formulations for Low/free Co active material

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Technical Accomplishments

- Assembled and tested 45 PPCs (2.1Ah), these cell were delivered to INL and TARDEC
- Surface coating using nano metal oxides on Hi-Ni NCMs enhances surface stability and improves cycle life
- CCA blends improved electrode conductivity and cell performance
- Surface modification and optimization of CCA formulation have led to increase in energy density and enhanced cell cycle life

Future Work

BP3 (2021-22)

- Optimization of Low-Co compositions to meet performance targets
- Select final surface modification process/composition for PCC selected cathode active material
- Optimize electrode formulation and architecture
- Build/test/deliver project Completion Cells: > 600 Wh/kg (cathode) after 1000 cycles with ≤ 5mg_{Co}/Wh CAM